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Spin-spin correlations in $Yb_2Ti_2O_7$: A polarized neutron scattering study

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Polarized neutron diffraction and neutron spin echo have been employed to investigate the low-temperature ($T < 2$ K) magnetic correlations in the frustrated magnet $Yb_2Ti_2O_7$. Several studies have reported a phase transition at 240 mK, however, the low-temperature phase is still under debate. It has been reported that $Yb_2Ti_2O_7$ enters a frozen ferromagnetic phase, however, studies of the spin dynamics suggest otherwise. Our results conclusively rule out a frozen ferromagnetic state and confirm that the majority of the spin system continues to fluctuate below 240 mK.

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Frustration, a condition in which competing interactions cannot be universally satisfied, is common in biology, chemistry, and physics.[1,2] Notable examples are protein folding, neural networks, and glasses, however, they are not well understood. Magnetic systems offer many examples of frustration, and one distinct class that has been studied extensively over the past decade is *geometrical frustration*,[3] where the spatial arrangement of atoms (geometry) competes with the other interactions between atomic spins (exchange, dipole, local anisotropy, etc.).

Theoretical arguments for antiferromagnetically coupled spins with both discrete[4] and continuous[5] symmetries on a lattice based on triangles have been discussed for some time. More recent work[6–8] also suggests that these systems do not enter a long-range Néel state but possess strong, short-range spatial correlations at finite temperatures. In reality, such magnets are particularly rare since, like the chemical lattice, the magnetic moments in materials usually freeze into ordered, or sometimes disordered, structures at a finite temperature.

Ytterbium titanate, $Yb_2Ti_2O_7$, is an insulator that crystallizes into the cubic pyrochlore structure[9] with the lattice parameter $a = 10.026(1)$ Å at room temperature (see Fig. 1).[10] It was believed to order just above 0.2 K,[11] where a sharp anomaly in the specific heat was observed. Hodges *et al.*[12] used Mössbauer spectroscopy to investigate the crystal field scheme and determined that the ground-state Kramers doublet was separated by 620 K from the first excited state, producing an easy plane anisotropy. They also found the effective paramagnetic moment to be $3.05(8)\mu_B$.

In a detailed magnetization study, Bramwell *et al.*[13] found a Curie-Weiss temperature of 0.59(1) K, indicative of weak ferromagnetic coupling, and a free ion moment of $3.335(4)\mu_B$, both consistent with earlier results.[12] Hodges and co-workers[14] extended their study of $Yb_2Ti_2O_7$ and found an abrupt change in the fluctuation rate of the Yb^{3+} spin at 0.24 K but not a frozen ground state as expected from the earlier studies.[11] Using muon spin relaxation and Mössbauer spectroscopies, they concluded that the Yb^{3+} spin fluctuations slow down by more than three orders of magnitude to several megahertz, without freezing completely. This was confirmed in their neutron powder diffraction, where no extra Bragg intensity was reported below 0.24 K. Recent single-crystal neutron diffraction by Yasui *et al.*[15] has revealed extra Bragg scattering below 0.24 K from a static ferromagnetic state, albeit with a reduced moment of $1.1\mu_B$. The latter two studies motivated our investigations.

Neutron spin echo (NSE) directly measures the intermediate scattering function, $S(\mathbf{Q},t)$, which contains information on both spatial and temporal spin correlations. NSE has been used in the past to study magnetic systems including spin glasses[16] and has recently been shown to be an ideal technique for studying the spin dynamics in geometrically frustrated systems.[17–19] The data obtained from such experiments are very sensitive to fluctuating spins with dynamics faster than $\sim 10^{-8}$ s, thus making it a probe of spin dynamics at relatively long times.

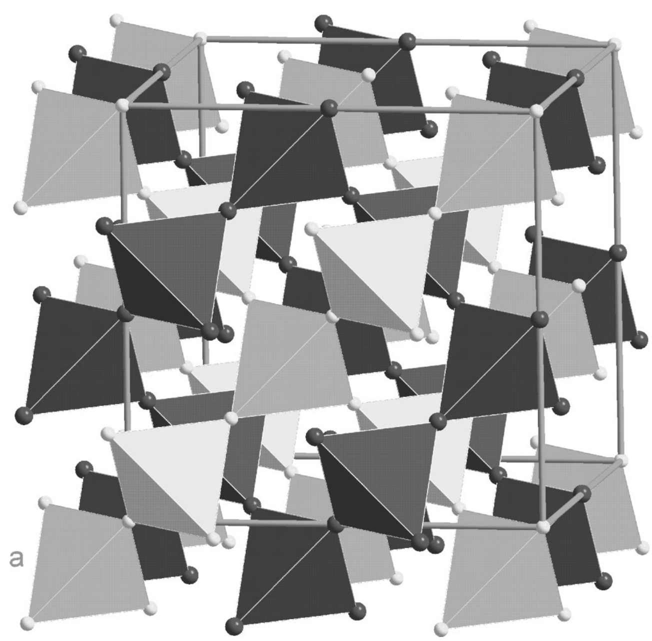

FIG. 1. The two-metal-ion sublattice that runs through the oxide pyrochlores. In $Yb_2Ti_2O_7$, only the Yb^{3+} has a magnetic moment.

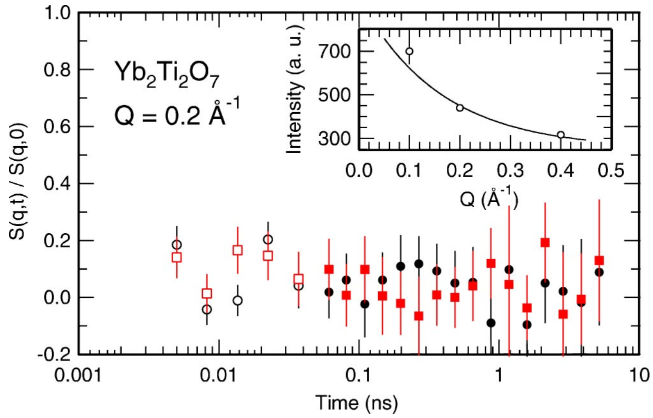

FIG. 2. (Color online) The normalized intermediate scattering function from $Yb_2Ti_2O_7$. Squares and circles are above (1 K) and below (180 mK) the 0.24 K transition, respectively. Open symbols use the shortened setup (see text). The signal completely relaxes in the first 4 ps. This is indicative of fast spin dynamics in a system, not unlike a paramagnet. The inset shows the Q dependence of the magnetic scattering as determined from *xyz* polarization analysis at 180 mK.

Polycrystalline $Yb_2Ti_2O_7$ was prepared by firing stoichiometric amounts of Yb_2O_3 and TiO_2 at 1350 °C for several days. The phase purity, room-temperature lattice parameter, and crystal structure of the samples were confirmed by x-ray diffraction. Static magnetization results are consistent, as are the structural parameters, with those published earlier. For the neutron scattering experiments, the sample was placed in a copper can and mounted on a dilution refrigerator for cooling down to 90 mK. Polarized neutron diffraction was performed on BT-2 with 13.7 meV neutrons and a small guide field (5 G) perpendicular to the scattering plane to maintain the beam polarization. The neutron spin-echo experiment was performed with an incident neutron beam of mean wavelength, $\lambda = 6.0$ Å and a 15% spread. The beam polarization was maintained by a 1 G field. NSE data were taken in the $|\mathbf{Q}|$ range between 0.1 and 0.4 Å^{-1}. This range in reciprocal space probes both short- and intermediate-range correlations. *xyz* polarization analysis was performed for each individual echo scan, at the same $|\mathbf{Q}|$, in order to relate the echo amplitude to the elastic magnetic-scattering intensity. This analysis separates magnetic scattering from coherent nuclear and incoherent spin and isotope scattering contributions that make up the total signal. A detailed description of magnetic spin echo is provided in Refs. 16 and 17. The echo measurement itself is only sensitive to magnetic scattering. The working dynamical range of the NSE spectrometer was extended by an order of magnitude towards lower correlation times by using a setup with shorter distance between the $\pi/2$ flippers, thus reducing the number of neutron spin precessions.

In agreement with Hodges,[14] our results confirm that the magnetic structure factor, $S(\mathbf{Q})$, at low $|\mathbf{Q}|$, increases as $|\mathbf{Q}|$ decreases. This is consistent with dominant ferromagnetic interactions (see inset of Fig. 2) and it is temperature independent above 0.18 K.

The normalized intermediate scattering function, $S(\mathbf{Q},t)/S(\mathbf{Q},0)$ at $|\mathbf{Q}| = 0.2$ Å^{-1}, is shown in Fig. 2 above (1 K) and below (0.18 K) the transition temperature seen by other techniques.[11,14,15] These measurements show that the signal relaxes before the NSE time window, that is within the first 4 ps. The two curves are within error of each other and all data points are within two sigma of zero (completely relaxed). This result is surprising since single-crystal diffraction studies[15] suggest $Yb_2Ti_2O_7$ is an ordered ferromagnet below 240 mK. A polycrystalline ferromagnet will generally depolarize the neutron beam when the internal domains are formed. However, the *xyz* polarization analysis confirmed the beam remained polarized throughout the experiment. Therefore, a long-range-ordered ferromagnetic phase below 0.24 K is inconsistent with the data presented here.

In an attempt to reconcile our results and those of the single-crystal experiment,[15] we have attempted to estimate the internal field and domain size. We have measured the polarization of the neutron beam transmitted by the sample and found that the change is less than 2% when going through the transition (conservative estimate). If P_0 is the incident beam polarization, then

$$\frac{P}{P_0} \approx \exp\left[-\frac{1}{2}\left(\frac{\gamma B}{v}\right)^2 R_d L\right], \tag{1}$$

where $\gamma/2\pi = -2916.4$ Hz/G is the gyromagnetic ratio of the neutron, B is the magnitude of the involved internal fields, v is the neutron velocity, R_d is the typical size of the magnetic domain, and L is the sample thickness.[20] With $\lambda = 6$ Å and $P/P_0 > 0.98$, we get an estimate of $B^2 R_d < 650$ G^2 μm. Assuming a $1\mu_B$ ordered moment, we estimate an unphysically small correlation length of 2 Å. On the other hand, if we assume a domain size of 1 μm, which is not unreasonable considering the resolution-limited Bragg scattering seen by Yasui *et al.*[15] the internal field is no more than 25 G or $0.01\mu_B$. The equation above applies to neutron depolarization due to *static* domains, but would also hold for correlated "units" fluctuating at 1 MHz (as suggested in Ref. 14), since this will appear static to a neutron traveling at 1 μm/ns.

Polarized neutron diffraction was performed at higher $|\mathbf{Q}|$'s to investigate the scattering around Bragg peaks. Neutrons with an incident energy of 13.7 meV were used and the spectrometer was in a double-axis configuration with a 60-min collimation before and after the sample. This data (shown in Fig. 3) confirms that $Yb_2Ti_2O_7$ does not depolarize the neutron beam and the presence of extra Bragg scattering at the (111) reciprocal lattice position. This Bragg scattering is clearly seen in the spin-flip channel, which conclusively identifies it as magnetic scattering. This is a very small amount of extra scattering and would be easily missed in an unpolarized powder experiment (as in Ref. 14), but it was seen by Yasui *et al.* because in their experiment the scattering was not powder averaged. We did not observe spin-flip scattering at other Bragg positions, probably as a result of the powder averaging. The amount of magnetic scattering at the (111) is consistent with the amount of magnetic scattering seen by Yasui *et al.* However, since we only observe one peak, we cannot calculate a structure factor (knowing the ferromagnetic structure proposed earlier is wrong) and a magnetic moment. With this data, however, we

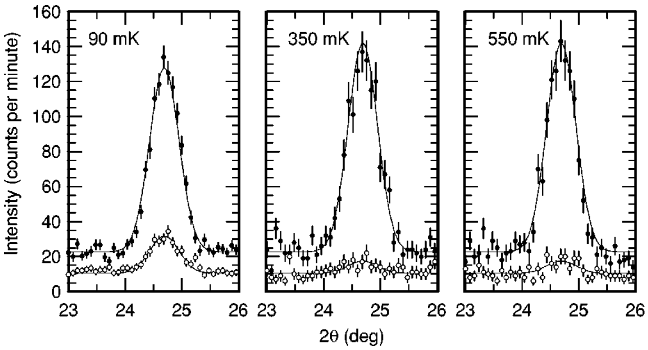

FIG. 3. Spin-flip (open circles) and non-spin-flip (closed circles) scattering at the (111) Bragg position. Above 240 mK there is only a small amount feed through due to incomplete polarization. At 90 mK, a peak is clearly seen in the spin-flip data.

can confirm the long-range nature of the correlations and discard the small domain scenario described above.

Two possibilities arise to explain the data presented here. First, a small fraction of the Yb spin system develops into a long-range, $q=0$ structure. This component cannot be ferromagnetic from the absence of depolarization, but it could be an antiferromagnetic structure or a more complicated structure. Unfortunately, without more data (preferably polarized single-crystal work) a magnetic structure cannot be proposed here. The data presented here will hopefully motivate others to perform such an experiment. The remaining spin system is dynamic and it is this component that causes the NSE to relax at all temperatures studied. Another possible explanation is that the small amount of spin-flip scattering observed in the diffraction experiment was due to the ordering of the nuclear spins. Such ordering would perturb the electronic spins and might result in the transition seen by muon and Mössbauer spectroscopy.

Whichever of the two scenarios is true, the interpretation of the spin-echo data is not affected, since it was measured at a different $|\mathbf{Q}|$. The majority of the Yb spin system remains dynamic below 240 mK with short-ranged ferromagnetic correlations.

This is consistent with the diffraction data by Hodges *et al.*, which implies a short-ranged correlation length of 20 Å (Fig. 1 in Ref. 14), but is inconsistent with their observed spin fluctuation rate of $\sim$1 MHz. Using Eq. (1) and a correlation length of 20 Å, this frequency yields a very low upper limit for the fluctuating fields ($\sim$1 G). With a spin-fluctuation rate of the order of 1 GHz, inferred from NSE, the sample would not be in the static limit, i.e., the internal fields could be larger without causing the neutrons to depolarize.

The $S(\mathbf{Q},t)/S(\mathbf{Q},0)$ observed here is very similar to that measured in the cooperative paramagnet, $Tb_2Ti_2O_7$.[19,21,22] NSE data from $Yb_2Ti_2O_7$ and $Tb_2Ti_2O_7$ (above 600 mK) are typical of spin relaxation normally observed in a paramagnet, where the system is too dynamic for this technique and uncorrelated spins completely relax the signal. To summarize, our studies on polycrystalline $Yb_2Ti_2O_7$ conclusively show this pyrochlore does not enter a *frozen* magnetic state at 240 mK. The data is also inconsistent with large ferromagnetic domains. In fact, on the NSE time scales, the system is dynamic above 180 mK.

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